Electro-diffusion in a plasma with two ion species

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Abstract

Electric field is a thermodynamic force that can drive collisional inter-ion-species transport in a multicomponent plasma. In an inertial confinement fusion (ICF) capsule, such transport causes fuel ion separation even with a target initially prepared to have equal number densities for the two fuel ion species. Unlike the baro-diffusion driven by ion pressure gradient and the thermo-diffusion driven by ion and electron temperature gradients, electro-diffusion has a critical dependence on the charge-to-mass ratio of the ion species. Specifically, it is shown here that electro-diffusion vanishes if the ion species have the same charge-to-mass ratio. An explicit expression for the electro-diffusion ratio is obtained and used to investigate the relative importance of electro- and baro-diffusion mechanisms. In particular, it is found that electro-diffusion reinforces baro-diffusion in the deuterium and tritium mix, but tends to cancel it in the deuterium and helium-3 mix.

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I. INTRODUCTION

In inertial confinement fusion (ICF) experiments, where the fuel assembly is a binary mixture of deuterium and tritium or deuterium and helium-3, the fusion power production is proportional to the product of number densities of the two species. Optimal fusion yield at a given target temperature in local thermodynamic equilibrium requires not only the fuel assembly be equi-molar for the two ion species, but also of equal number densities everywhere in the assembly [1]. In terms of the mass densities, this suggests that the mass density of the light ions

$$\rho_l = m_l n_l$$

should be m_l/m_h times of the heavy ion mass density

$$\rho_h = m_h n_h$$

for $n_l = n_h$. Here the subscript "l" denotes light ion species and "h" denotes heavy ion species. Defining the mass concentration of the light ions as

$$c \equiv \frac{\rho_l}{\rho}$$

with the mixture ion mass density

$$\rho \equiv \rho_l + \rho_h,$$

one finds that the optimal arrangement of equal ion number densities implies a spatially uniform $c = m_l/(m_l + m_h)$. This condition can be accurately satisfied in the initial target preparation. The dynamical process of implosion, however, can introduce light and heavy ion separation which degrades the fusion power production [2–4].

The collisional inter-ion-species transport or concentration diffusion is driven by the concentration gradient ∇c as well as other thermodynamic forces such as the ion pressure gradient, electron and ion temperature gradients, and electric field. In the case of a plasma with two species of ions, we show that the diffusive ion mass flux $\mathbf{i}$ takes the general form of

$$\mathbf{i} = -\rho D\Big(\nabla c + k_p \nabla \log p_i + \frac{e k_E}{T_i}\nabla\Phi + k_T^{(i)}\nabla \log T_i + k_T^{(e)}\nabla \log T_e\Big).$$

This flux governs c evolution through

$$\rho\frac{\partial c}{\partial t} + \rho\mathbf{u}\cdot\nabla c + \nabla\cdot\mathbf{i} = 0, \tag{1}$$

where $\mathbf{u}$ is the plasma fluid velocity.

For the ICF fuel assembly, even when the initial condition has $\nabla c = 0$ by design, the theromodynamic cross terms can drive significant diffusive flux $\mathbf{i}$ through the baro-diffusion ($k_p \neq 0$), electro-diffusion ($k_E \neq 0$), and thermo-diffusion ($k_T^{(e,i)} \neq 0$). The above equation attributes baro-diffusion to the total ion pressure gradient ∇p_i, which is the sum of the ion species pressure gradients. This is different from the case of the neutral gas mixture, where baro-diffusion is considered to be due to the total mixture pressure. Also, because of the large difference between the electron and ion masses their temperatures can vary. In the general case, thermo-diffusion is driven by gradients of both the electron and ion temperatures.

Baro-diffusion is fundamentally the result of the mass depedence of the species thermal speed, and we will show it to be indepedent of electric field and effective gravity due to acceleration and decceleration in implosions of the ICF target, just like its counterpart in neutral gas mixtures [5, 6]. This finding should be contrasted with a previous result [2, 3] which suggests gravity and electric field dependence of k_p for binary plasma mixture. The role of electric field, or electro-diffusion, is a feature intrinsic to a plasma. It is fundamentally the result of the different acceleration experienced by the ions of different charge-to-mass ratio in an electric field. In a low temperature plasma with significant neutral gas background, this effect is known as ions having different mobilities [7]. We show that electro-diffusion vanishes ($k_E = 0$) if the charge-to-mass ratio is identical for the two otherwise distinct ion species. The charge-to-mass ratio dependence of k_E allows drastically different electro-diffusion behavior for different binary plasma mixtures. For the DT mix, we find that

$$k_E = k_p,$$

so the electro- and baro-diffusion reinforce each other in a plasma shock. In contrast, for the D^3He mix, one has

$$k_E = -k_p,$$

which implies that electro-diffusion tends to cancel baro-diffusion in a plasma shock.

The thermo-diffusion is fundamentally the result of the thermal force in the collisional drag between different ion species and between electrons and ions. The ion-ion thermal force produces a non-vanishing $k_T^{(i)}$, while the ion-electron thermal force leads to a finite $k_T^{(e)}$. Because of these dependences, thermo-diffusion coefficients require a kinetic calculation

of the said transport coefficients through the distribution function perturbed from a local Maxwellian. Hence, $k_T^{(i,e)}$ are not thermodynamic quantities like k_p and k_E. It must be noted that with comparable ion masses, the thermal force evaluation is a much more involved exercise, than the electron-ion one carried out by Braginskii [8]. The critical information is nevertheless implicitly contained in standard transport calculations for multi-component plasmas such as that by Hirshman and Sigmar [9]. Explicit evaluation of the thermo-diffusion coefficients for the DT and D^3He mixtures will be carried out in a future work.

The inter-ion-species diffusion, which modifies the relative number density of the two fusion reactants, has been an issue of interest in inertial confinement fusion and dense plasma research. Among those we are familiar with, C. H. Chang, B. Albright, and W. Daughton from Los Alamos National Laboratory have investigated the models for $\mathbf{i}$ with varying degrees of approximation for evaluating the baro- and thermo-diffusion in the past decade, albeit in unpublished reports. More recently, it has been attracting special attention for the pioneering analysis of Amendt et al [2, 3], which shows baro-diffusion may be responsible for the discrepancy between the neutron yield measured during ICF implosions and that predicted by simulations. This can largely be attributed to the realization that the strong pressure and temperature gradients, along with the strong electric field [10], are induced by shock waves inevitably present in the imploded capsule. Inter-diffusion between the two ion species must therefore take place; the resulting separation of the fuel constituents in the hot spot can significantly degrade the fusion yield [2, 3]. An experimental evidence for the fuel stratification in ICF implosions has been reported by Casey et al based on their measurements at the OMEGA laser facility [11].

The purpose of this paper is to clarify the underlying diffusion mechanisms, especially the role of electro-diffusion in relation to baro-diffusion. The derivation of $\mathbf{i}$ based on multi-component collisional fluid models also provides a framework for incorporating this important physics in ICF modeling. As highlighted earlier for a binary plasma mix, there are a number of subtleties absent in neutral gas mixture, and not addressed in preceding studies. Since the inter-species diffusion is a general topic for multi-component plasmas, it is expected that the results described here would be of general interest, for example, to tokamak edge plasma modeling, plasma processing, and stellar structures.

The rest of the paper is organized as follows. In the next section, the thermodynamic framework for evaluating electro-diffusion is outlined. In section III, the momentum equation

for an ion species in the center-of-mass frame is obtained and simplified by imposing an ordering relevant to a collisional plasma shock. Then, in section IV, this equation is utilized to write the diffusive mass flux of an ion species in the general form of section II, thereby explicitly evaluating the electro-diffusion ratio. Finally, in section V, implications of electro-diffusion in the ICF context are discussed.

II. THERMODYNAMIC EXPRESSION FOR THE DIFFUSIVE FLUX

To define the diffusive mass flux of ion component α, the center-of-mass velocity $\mathbf{u}$ is first introduced by

$$\rho\mathbf{u} \equiv \sum_{\alpha} \rho_\alpha \mathbf{u}_\alpha, \tag{2}$$

where ρ_α is the partial density of component α and $\rho = \sum_\alpha \rho_\alpha$ is the mixture total ion density. Also, $\mathbf{u}_\alpha$ denotes the net flow velocity of the component and the sum on the right side of Eq. (2) is over all the components present in the mix. The diffusive mass flux is then given by

$$\mathbf{i}_\alpha = \rho_\alpha(\mathbf{u}_\alpha - \mathbf{u}). \tag{3}$$

If the system is close to local thermodynamic equilibrium, a linear relation between the thermodynamic forces and the resulting fluxes can be assumed. For a neutral gas mixture, the total diffusive mass flux of component α can be written as [5, 6]

$$\mathbf{i}_\alpha = -\rho D\Big(\nabla c_\alpha + k_p \nabla \log p + k_T \nabla \log T\Big), \tag{4}$$

where p and T denote the mixture total pressure and temperature, respectively, and $c_\alpha \equiv \rho_\alpha/\rho$ denotes concentration of the component α. Parameter D is called diffusion coefficient; it governs the diffusive flux when only the concentration gradient is present. In view of Eq. (4), baro- and thermo-diffusion coefficients are then equal to $k_p D$ and $k_T D$, respectively. Dimensionless parameters k_p and k_T are usually referred to as baro- and thermo-diffusion ratios, respectively.

Interestingly, k_p is a thermodynamic quantity, i.e. it can be evaluated given local values of thermodynamic variables and does not depend on the details of collisions [5]. In particular, for a binary mix it can be found [5, 6]

$$k_p = c(1-c)(m_h - m_l)\Big(\frac{c}{m_l} + \frac{1-c}{m_h}\Big), \tag{5}$$

where m_l and m_h are the atomic masses of the light and heavy fractions, respectively. Also, $c \equiv c_l$ stands for the concentration of the light fraction; the subscript "l" is dropped to simplify notation, as in a binary mix the concentration of the heavy fraction can be recovered through $c_h = 1 - c_l$. In contrast to k_p, k_T is an intrinsically kinetic quantity and is subject to change depending on the collisional model.

To account for the effect of the electric field, the corresponding force needs to be added on the right side of Eq. (4) to rewrite it as

$$\mathbf{i}_\alpha = -\rho D \Big(\nabla c_\alpha + k_p \nabla \log p + k_T \nabla \log T + \frac{e k_E}{T} \nabla \Phi \Big), \tag{6}$$

where Φ is the electrostatic potential and, by analogy with k_p and k_T, the electro-diffusion ratio k_E is introduced. In what follows, we focus on the case of a binary plasma mix, i.e. plasma consisting of two ion species and electrons. To illuminate the new features brought by the electric field, as compared to the case of a binary mix of neutral gases, we apply Eq. (6) to the system consisting of the two ion species. Within such an approach, the electron species is viewed as an external factor that affects the system of interest through the electric field and collisions. In other words, the electrons do not make contribution in the definition of $\rho, \mathbf{u}$, and p, that is consistent with our objective to understand the relative motion of the two ion species. It is worth noticing that because of small inertia the electron contribution to the overall plasma density and flow is negligible. Thus, for all practical purposes ρ and $\mathbf{u}$ can still be referred to as the plasma density and flow, respectively. On the contrary, the electron and ion pressures are generally comparable and employing the total ion pressure, rather than the overall plasma pressure, in place of p in Eq. (6) is crucial.

Assuming the diffusive flux of the form (6) it is possible to evaluate k_E by generalizing formal thermodynamic methods used in Ref. [5] to evaluate k_p [4]. Instead, here we start from the first-principle based momentum conservation equations for individual species to automatically recover this form. Importantly, in addition to readily providing k_p and k_E, this technique gives formulae for D, $k_T^{(i)}$ and $k_T^{(e)}$ in terms of standard transport coefficients. In so doing, it lays the framework for evaluating the overall effect of the ion concentration diffusion that is inherently not possible within the thermodynamic approach.

III. MOMENTUM CONSERVATION FOR ION SPECIES

We start by writing the momentum equations for the two ion species

$$\rho_\alpha \frac{d_\alpha \mathbf{u}_\alpha}{dt} + \nabla \cdot \overleftrightarrow{P}_\alpha - n_\alpha Z_\alpha e \mathbf{E} - \rho_\alpha \mathbf{F}_\alpha = \sum_\beta \mathbf{R}_{\alpha\beta}, \tag{7}$$

where the subscript α can be "l" and "h" to denote the light and heavy ion species, respectively. In Eq. (7) n_α, p_α and Z_α stand for the species' number density, partial pressure and charge number, respectively. Also, $\mathbf{E}$ stands for the electric field. Acceleration due to an external force of a non-electric origin, such as the gravitational force, is denoted by $\mathbf{F}_\alpha$, while $\mathbf{R}_{\alpha\beta}$ is the force density due to collisional momentum exchange with the species β and the sum on the right side of Eq. (7) is over all plasma species, including electrons. The pressure tensor $\overleftrightarrow{P}_\alpha$ is defined in the frame co-moving with the species net flow by

$$\overleftrightarrow{P}_\alpha \equiv m_\alpha \int (\mathbf{v} - \mathbf{u}_\alpha)(\mathbf{v} - \mathbf{u}_\alpha) f_\alpha d^3 v,$$

where f_α is the species distribution function. Finally,

$$d_\alpha/dt \equiv \partial/\partial t + \mathbf{u}_\alpha \cdot \nabla.$$

Next, we obtain the momentum equation for the center of mass velocity defined by Eq. (2). To do so, it is convenient to rewrite Eq. (7) in the conservative form:

$$\frac{\partial(\rho_\alpha \mathbf{u}_\alpha)}{\partial t} + \nabla \cdot (\rho_\alpha \mathbf{u}_\alpha \mathbf{u}_\alpha) + \nabla \cdot \overleftrightarrow{P}_\alpha - n_\alpha Z_\alpha e \mathbf{E} - \rho_\alpha \mathbf{F}_\alpha = \sum_\beta \mathbf{R}_{\alpha\beta}, \tag{8}$$

where species continuity equation

$$d_\alpha \rho_\alpha / dt + \rho_\alpha \nabla \cdot \mathbf{u}_\alpha = 0$$

is used. Introducing the species' velocity in the center-of-mass frame

$$\mathbf{w}_\alpha \equiv \mathbf{u}_\alpha - \mathbf{u}, \tag{9}$$

noticing that

$$\nabla \cdot (\rho_\alpha \mathbf{u}_\alpha \mathbf{u}_\alpha) = \nabla \cdot (\rho_\alpha \mathbf{u}\mathbf{u}) + \nabla \cdot (\rho_\alpha \mathbf{w}_\alpha \mathbf{u}) + \nabla \cdot (\rho_\alpha \mathbf{u} \mathbf{w}_\alpha) + \nabla \cdot (\rho_\alpha \mathbf{w}_\alpha \mathbf{w}_\alpha)$$

and summing Eq. (8) over all the ion species we find

$$\frac{\partial(\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u}\mathbf{u}) + \nabla \cdot \overleftrightarrow{P}_i - \sum_{\alpha = i_j} (n_\alpha Z_\alpha e \mathbf{E} + \rho_\alpha \mathbf{F}_\alpha) = \sum_{\alpha = i_j} \mathbf{R}_{\alpha e}, \tag{10}$$

where we use that $\sum_{\alpha=i_j} \rho_\alpha \mathbf{w}_\alpha = 0$, $\overleftrightarrow{P}_i = \sum_\alpha(\overleftrightarrow{P}_\alpha + \rho_\alpha \mathbf{w}_\alpha \mathbf{w}_\alpha)$ is the total ion pressure tensor in the center-of-mass frame and subscript "$\alpha = i_j$" denotes summation over the ion species only. Equation (10) is then easy to transform to a more familiar form

$$\rho \frac{d\mathbf{u}}{dt} + \nabla p_i + \nabla \cdot \overleftrightarrow{\Pi}_i - \sum_{\alpha=i_j} (n_\alpha Z_\alpha e \mathbf{E} + \rho_\alpha \mathbf{F}_\alpha + \mathbf{R}_{\alpha e}) = 0, \tag{11}$$

where $d/dt \equiv \partial/\partial t + \mathbf{u} \cdot \nabla$ and p_i and $\overleftrightarrow{\Pi}_i$ are the total ion pressure and viscous stress tensor, respectively.

Finally, we rewrite equations for the individual species flows in the center-of-mass frame to obtain

$$\frac{d\rho_\alpha \mathbf{w}_\alpha}{dt} + \rho_\alpha \mathbf{w}_\alpha \cdot \nabla \mathbf{u} + \rho_\alpha \mathbf{w}_\alpha \nabla \cdot \mathbf{u} + \nabla \cdot \overleftrightarrow{P}_\alpha^{c.m} - n_\alpha Z_\alpha e \mathbf{E} - \rho_\alpha \mathbf{F}_\alpha + \rho_\alpha \frac{d\mathbf{u}}{dt} = \sum_\beta \mathbf{R}_{\alpha\beta}, \tag{12}$$

where

$$\overleftrightarrow{P}_\alpha^{c.m} = \overleftrightarrow{P}_\alpha + \rho_\alpha \mathbf{w}_\alpha \mathbf{w}_\alpha = m_\alpha \int (\mathbf{v} - \mathbf{u})(\mathbf{v} - \mathbf{u}) f_\alpha d^3 v$$

is the species' pressure tensor in the center-of-mass frame. By splitting a scalar pressure out of $\overleftrightarrow{P}_\alpha^{c.m}$, Eq. (12) is then rewritten further to find

$$\frac{d\rho_\alpha \mathbf{w}_\alpha}{dt} + \rho_\alpha \mathbf{w}_\alpha \cdot \nabla \mathbf{u} + \rho_\alpha \mathbf{w}_\alpha \nabla \cdot \mathbf{u} + \nabla p_\alpha^{c.m} + \nabla \cdot \overleftrightarrow{\Pi}_\alpha^{c.m} - n_\alpha Z_\alpha e \mathbf{E} - \rho_\alpha \mathbf{F}_\alpha + \rho_\alpha \frac{d\mathbf{u}}{dt} = \sum_\beta \mathbf{R}_{\alpha\beta}, \tag{13}$$

where $p_\alpha^{c.m}$ and $\overleftrightarrow{\Pi}_\alpha^{c.m}$ are the species' partial pressure and viscous stress tensor, respectively, evaluated in the center-of-mass frame.

The calculation presented here is motivated by the problem of the ion species diffusion within an ICF relevant shock wave front. In general, the shock front width can be as small as the mean free path, making a local treatment, as well as the framework of the previous section, invalid. However, for a moderately strong shock, the front width can be assumed much greater than the mean free path λ, i.e.

$$\lambda/\Delta << 1, \tag{14}$$

where Δ is the characteristic spatial scale of the plasma (e.g. the shock width). The mean free paths can substantially differ for pre- and post-shocked plasmas; for definitiveness, we refer λ to the post-shock mean free path. Ordering (14) is usually satisfied for Mach numbers $\lesssim 2$ and ensures that the plasma remains mostly collisional throughout the shock front [12]. The characteristic temporal scale τ can then be estimated from $\tau^{-1} \sim v_{sh}/\Delta$, where v_{sh}

is the shock speed. Assuming that ion masses are comparable across different species and introducing the characteristic ion thermal speed $v_{th-i} \sim v_{sh}$ we find

$$\nu_i \tau \sim (\lambda/\Delta)^{-1} >> 1, \tag{15}$$

where $\nu_i \sim v_{th-i}/\lambda$ is the characteristic ion collision frequency.

Estimating the friction between the ion species by $\mu_{lh}\nu_i n_l(w_l - w_h)$, where μ_{lh} is the reduced mass for the light and heavy ions, it is straightforward to show that ordering (15) makes

$$\frac{w_\alpha}{v_{th-i}} \sim \frac{\lambda}{\Delta} \ll 1,$$

thereby ensuring that the system is close to a local equilibrium. The terms on the left side of Eq. (13) that contain both the spatial gradient and w_α are quadratic in the small parameter λ/Δ and can be dropped. For the same reason $\overleftrightarrow{P}^{c.m}_\alpha \approx \overleftrightarrow{P}_\alpha$ and the superscript "$c.m.$" appearing next to the partial pressure and viscous tensor can be omitted. Finally, due to the same estimate for the friction, the $\partial \mathbf{w}_\alpha/\partial t$ term can be dropped as well and Eq. (13) reduces to

$$\nabla p_\alpha + \nabla \cdot \overleftrightarrow{\Pi}_\alpha - n_\alpha Z_\alpha e \mathbf{E} - \rho_\alpha \mathbf{F}_\alpha + \rho_\alpha \frac{d\mathbf{u}}{dt} = \sum_\beta \mathbf{R}_{\alpha\beta}. \tag{16}$$

Equation (16) is valid for a plasma with an arbitrary number of species as long as ordering (15) is obeyed. In the next section, we apply it to evaluate the electro-diffusion coefficient in a plasma with two ion species.

IV. EVALUATING ELECTRO-DIFFUSION COEFFICIENT

The viscous term appearing on the left side of Eq. (16) is governed by the second order derivatives of macroscopic parameters. Its contribution is therefore not retained in Eq. (6), which is obtained by assuming linear relation between the thermodynamic forces and the resulting flux. In principle, this contribution may be substantial and effectively modify the baro-diffusion ratio [5, 6]. However, as the main goal of the present study is to elucidate the role of the electric field on the ion diffusion, in what follows we drop $\nabla \cdot \overleftrightarrow{\Pi}_\alpha$ on the left side of Eq. (16). Then, employing Eq. (11) with the $\nabla \cdot \overleftrightarrow{\Pi}_i$ term also dropped to evaluate du/dt in Eq. (16) we find

$$\sum_{\beta=i_j} \mathbf{R}_{\alpha\beta} + (\mathbf{R}_{\alpha e} - \frac{\rho_\alpha}{\rho} \sum_{\beta=i_j} \mathbf{R}_{\beta e}) =$$

$$(\nabla p_\alpha - \frac{\rho_\alpha}{\rho}\nabla p_i) - (Z_\alpha n_\alpha - \frac{\rho_\alpha}{\rho}\sum_{\beta=i_j} Z_\beta n_\beta)e\mathbf{E} - (\rho_\alpha \mathbf{F}_\alpha - \frac{\rho_\alpha}{\rho}\sum_{\beta=i_j}\rho_\beta \mathbf{F}_\beta). \tag{17}$$

Notice, that if we had included electrons into the system, the sum over β in the second term on the right side of Eq. (17) would vanish due to quasi-neutrality.

Equation (17) gives the light ion species diffusion velocity $\mathbf{w}_l$ through the $\mathbf{R}_{lh}$ dependence on the net velocity difference between the ion species since

$$\mathbf{w}_l - \mathbf{w}_h = \Big(1 + \frac{\rho_l}{\rho_h}\Big)\mathbf{w}_l = \frac{\mathbf{w}_l}{1-c}, \tag{18}$$

where c is the concentration of the light ion species. In a multi-component plasma [9], the collisional drag between species α and β

$$\mathbf{R}_{\alpha\beta} = -[A_{\alpha\beta}\mu_{\alpha\beta}n_\alpha\nu_{\alpha\beta}(\mathbf{w}_\alpha - \mathbf{w}_\beta) + c^{(1)}_{\alpha\beta}n_\alpha\nabla T_\alpha + c^{(2)}_{\alpha\beta}n_\beta\nabla T_\beta], \tag{19}$$

where, in general, coefficients $A_{\alpha\beta}$, $c^{(1)}_{\alpha\beta}$ and $c^{(2)}_{\alpha\beta}$ are complicated functions of the masses, densities and charge numbers of all the species and $\mathbf{R}_{\alpha\alpha} = 0$ for arbitrary T_α implies $c^{(1)}_{\alpha\alpha} = c^{(2)}_{\alpha\alpha} = 0$. Also, T_α is the temperature of species α, $\mu_{\alpha\beta} \equiv m_\alpha m_\beta/(m_\alpha + m_\beta)$ is the reduced mass and $\nu_{\alpha\beta}$ stands for the frequency of collisions between species α and β. Conventionally, the terms on the right side of Eq. (19) proportional to the velocity difference and temperature gradients are referred to as the frictional and thermal forces, respectively. Summing Eq. (19) over the ion species we find

$$\sum_{\beta=i_j}\mathbf{R}_{\alpha\beta} = -\sum_{\beta=i_j}[A_{\alpha\beta}\mu_{\alpha\beta}n_\alpha\nu_{\alpha\beta}(\mathbf{w}_\alpha - \mathbf{w}_\beta) + B_{\alpha\beta}n_\beta\nabla T_\beta], \tag{20}$$

where $B_{\alpha\alpha} \equiv \sum_{\beta=i_j} c^{(1)}_{\alpha\beta}$ and $B_{\alpha\beta} \equiv c^{(2)}_{\alpha\beta}$ for $\alpha \neq \beta$.

When the elementary masses of species α and β are comparable, the thermal force acting between them depends on both ∇T_α and ∇T_β, i.e. $c^{(1)}_{\alpha\beta} \sim c^{(2)}_{\alpha\beta}$ on the right side of Eq. (19). In contrast, the thermal force acting between the electron and any of the ion species is dominated by the electron temperature gradient, because the thermal speed of electrons is much greater than that of ions, *i.e.*,

$$\mathbf{R}_{\alpha e} = -[A_{\alpha e}\mu_{\alpha e}n_\alpha\nu_{\alpha e}(\mathbf{w}_\alpha - \mathbf{w}_e) + B_{\alpha e}n_e\nabla T_e] \tag{21}$$

where we set $B_{\alpha e} \equiv c^{(2)}_{\alpha e}$ to unify notation with Eq. (20), and

$$\mathbf{w}_e \equiv \mathbf{u}_e - \mathbf{u}$$

is the electron flow velocity in the ion center-of-mass flow frame. Unlike $\mathbf{w}_{l,h} \sim (\lambda/\Delta)v_{th-i}$ from our collisional ordering, there is no such constraint on the electron flow $\mathbf{w}_e$ due to the much larger electron thermal velocity. This is consistent with the well-known result that a collisional plasma can carry a substantial current in the electron channel despite that the ion current is negligibly small in the short mean-free-path limit. To estimate the ion-electron frictional force on the right side of Eq. (21) we introduce the plasma current

$$\mathbf{J} \equiv -en_e\mathbf{u}_e + \sum_{\beta=i_j} en_\alpha Z_\alpha \mathbf{u}_\alpha = -en_e\mathbf{w}_e + \sum_{\beta=i_j} en_\alpha Z_\alpha \mathbf{w}_\alpha, \tag{22}$$

where the quasi-neutrality condition along with Eq. (9) is used to obtain the right side of the equation. Then, using Eq. (18), the friction between the light ions and electrons can be rewritten as

$$\mathbf{R}^f_{le} = -A_{le}\mu_{le}n_l\nu_{le}(\mathbf{w}_l - \mathbf{w}_e) = -A_{le}\mu_{le}n_l\nu_{le}\Big[\frac{Z_h/m_h}{cZ_l/m_l + (1-c)Z_h/m_h}\mathbf{w}_l + \frac{1}{en_e}\mathbf{J}\Big]. \tag{23}$$

The term on the right side involving $\mathbf{w}_l$ is smaller than the friction between the ion species by a factor of $\sqrt{m_e/m_{l,h}}$. In an ambipolar plasma $\mathbf{J} = 0$ and the frictional force can be neglected on the right side of Eq. (21). Moreover, even for a plasma carrying significant current through the electrons due to the $-en_e\mathbf{w}_e$ term in Eq. (22), the ion-electron friction force is much less than its ion-ion counterpart as long as

$$\frac{J}{en_e v_{th-i}} \ll \frac{\lambda}{\Delta}\sqrt{\frac{m_{l,h}}{m_e}}. \tag{24}$$

Condition (24) is the most restrictive in the case of a weak shock, where the shock front width can be many times of the ion-ion mean free path making $(\lambda/\Delta)\sqrt{m_{l,h}/m_e}$ of order unity [12]. The constraint on the plasma current becomes $J \ll en_e v_{th-i}$. Hence, in the absence of large currents on the order of $en_e v_{th-i}$ or greater, $\mathbf{R}^f_{\alpha e}$ can be ignored for an ICF relevant shock wave. Consequently, the ion-electron collisional drag is dominated by the thermal force,

$$\mathbf{R}_{\alpha e} \approx -B_{\alpha e}n_e\nabla T_e. \tag{25}$$

Applying general expressions (20) and (25) to our case and setting $\alpha = l$ the left side of Eq. (17) is now evaluated to find

$$\sum_{\beta=i_j}\mathbf{R}_{l\beta} + (\mathbf{R}_{le} - \frac{\rho_l}{\rho}\sum_{\beta=i_j}\mathbf{R}_{\beta e}) =$$
$$-A_{lh}\mu_{lh}n_l\nu_{lh}(\mathbf{w}_l - \mathbf{w}_h) - \sum_{\beta=l,h} B_{l\beta}n_\beta\nabla T_\beta - (B_{le} - c\sum_{\beta=l,h}B_{\beta e})n_e\nabla T_e. \tag{26}$$

While the coefficients $B_{\beta e}$ are relatively easy to recover by generalizing the corresponding Braginskii's result for a simple plasma [8], evaluating A_{lh}, B_{lh} and B_{ll} is quite complicated even in the case of only two different ion species with comparable masses, charge numbers and concentrations. Fortunately, it will be found unnecessary for the purpose of this paper, so we proceed leaving coefficients of Eq. (26) unspecified.

To complete the calculation, Eq. (17) needs to be rewritten in the canonical form (6). The terms on the right side of Eq. (17) then have to be expressed in terms of the total ion pressure p_i and the light species concentration c. We now proceed by doing so in the first term on the right side of Eq. (17) that is responsible for baro-diffusion.

First, we observe that energy exchange *between* the ion species with comparable masses takes place over the same time scale as thermal equilibration *within* any of the two species. Hence, under ordering (15), $T_l \approx T_h$ and the overall ion temperature T_i can be introduced. Next, we notice that $p_i = (n_l + n_h)T_i = (\rho_l/m_l + \rho_h/m_h)T_i$, where m_l and m_h are the light and heavy ion masses, respectively, to obtain

$$p_l = \frac{cm_h}{cm_h + (1-c)m_l} p_i, \tag{27}$$

and

$$\nabla p_l = \frac{cm_h}{cm_h + (1-c)m_l} \nabla p_i + \frac{p_i m_h m_l}{[cm_h + (1-c)m_l]^2} \nabla c. \tag{28}$$

The first term on the right side of Eq. (17) is then evaluated to find

$$(\nabla p_l - \frac{\rho_l}{\rho} \nabla p_i) = \frac{\rho T_i}{cm_h + (1-c)m_l} \Big[\nabla c + c(1-c)(m_h - m_l)\Big(\frac{c}{m_l} + \frac{1-c}{m_h}\Big) \nabla \log p_i\Big]. \tag{29}$$

Expression inside the square brackets of Eq. (29) is normalized, i.e. the coefficient in front of the ∇c term is equal to unity. In view of Eq. (6) it means that the coefficient in front of the $\nabla \log p_i$ term is equal to the baro-diffusion ratio k_p. Importantly, this ratio, obtained here from ion fluid equations, matches the result (5), found in Refs. [5, 6] for a binary mix of ideal gases. Of course, this is just a reflection of the aforementioned fact that k_p is a thermodynamic quantity and does not depend on the details of the collisional exchange between the species. It should be noted that recovering the same k_p as in Refs. [5, 6] manifests the key difference between our approach and that of Refs. [2, 3], where k_p is found to be dependent upon the electric field.

With the technique presented in the preceding paragraphs, k_E can be straightforwardly calculated in the same way as k_p. Before doing so, we take a brief detour and apply this

technique to clarify the role of gravity. The effective gravity appears in ICF relevant problems when acceleration of the capsule during implosion needs to be accounted for. Upon switching to the frame co-moving with the capsule the inertial force enters the momentum equation that is formally equivalent to placing the system into external field with an effective gravitational acceleration $\mathbf{g}$.

In Refs. [2, 3], gravity is found to modify the expression for k_p, so does the electric field. Within the framework of the present study, gravity can be included by setting the external force $\mathbf{F}_\alpha$ equal to $\mathbf{g}$ for both $\alpha = l$ and $\alpha = h$. The third term on the right side of Eq. (17) is then found to vanish; that is, the gravitational force does not drive a diffusive flux. This result obtained with a rather formal method has a trivial physical explanation. Namely, gravity gives the same acceleration to all ions regardless of their mass and charge number and therefore introducing it into otherwise unchanged system does not directly contribute to the species separation. Of course, gravity can still affect ion concentrations indirectly. For example, it can do so by modifying the electron pressure balance. The electric field then has to adjust, thereby modifying the ion flux through its electro-diffusive component.

Now we obtain the electro-diffusion ratio by writing the total diffusive flux of the light ion species in the canonical form. The second term on the right side of Eq. (17) is first evaluated to find

$$(Z_l n_l - \frac{\rho_l}{\rho} \sum_{\beta = i_j} Z_\beta n_\beta) e\mathbf{E} = \rho c(1-c)\Big(\frac{Z_l}{m_l} - \frac{Z_h}{m_h}\Big) e\mathbf{E}. \tag{30}$$

Next, the terms on the right side of Eq. (17) are collected with the help of Eqs. (29) and (30) and Eq. (26) is employed along with Eq. (18) to find

$$\mathbf{i}_l \equiv \rho_l \mathbf{w}_l = -\rho D\Big(\nabla c + k_p \nabla \log p_i + \frac{e k_E}{T_i}\nabla\Phi + k_T^{(i)}\nabla \log T_i + k_T^{(e)}\nabla \log T_e\Big), \tag{31}$$

where, as recovered by Eq. (29), k_p is still given by Eq. (5) and

$$D = \frac{\rho T_i}{A_{lh}\mu_{lh} n_l \nu_{lh}} \times \frac{c(1-c)}{c m_h + (1-c) m_l}, \tag{32}$$

$$k_E = m_l m_h c(1-c)\Big(\frac{c}{m_l} + \frac{1-c}{m_h}\Big)\Big(\frac{Z_l}{m_l} - \frac{Z_h}{m_h}\Big), \tag{33}$$

$$k_T^{(i)} = m_l m_h\Big(\frac{c}{m_l} + \frac{1-c}{m_h}\Big)\Big[\frac{c B_{ll}}{m_l} + \frac{(1-c) B_{lh}}{m_h}\Big], \tag{34}$$

$$k_T^{(e)} = m_l m_h\Big(\frac{c}{m_l} + \frac{1-c}{m_h}\Big)\Big[\frac{c Z_l}{m_l} + \frac{(1-c) Z_h}{m_h}\Big][(1-c) B_{le} - c B_{he}]\frac{T_e}{T_i}, \tag{35}$$

where quasi-neutrality condition was used to write Eq. (35).

Expression (31) does not have the exact form of Eq. (6) since the $\nabla \log T_e$ term appears on the right side. This is because the only external force accounted for by Eq. (6) is the electric field, whereas for the system of the two ion species considered here the thermal force exerted by electrons is also external. Moreover, equation (6) is only valid when at any given point different components of the system are nearly equilibrated; in particular, this means that temperatures of all the components must be equal. For the system including ions only, this condition is satisfied due to our ordering (15). However, this ordering does allow T_e to be different from T_i, as the energy exchange between the electron and any of the ion species takes longer than that between the two ion species by a factor of $\sqrt{m_{l,h}/m_e}$. Hence, even for the plasma as a whole, for which the ion-electron thermal force is internal, T_e and T_i have to be set equal for thermodynamically obtained Eq. (6) to be recovered. It is interesting to note that $T_i \neq T_e$ is normally expected in an ICF capsule, especially at the hot spot where fusion occurs.

Equations (32) and (33) give the electro-diffusion coefficient $k_E D$, thereby fulfilling the goal of this paper. Notice that k_E goes to zero if the charge-to-mass ratios are equal for the two ion species. This result rigorously obtained here from ion fluid equations has a simple physical explanation. Indeed, when $Z_l/m_l = Z_h/m_h$ the electric field does not distinguish between the light and heavy ions and therefore does not contribute to the relative motion of the species.

Unlike expressions (34) - (35) for thermo-diffusion ratios, which involve transport coefficients $B_{\alpha\beta}$, Eq. (33) provides an explicit result for k_E without invoking a kinetic calculation. In other words, as its baro-diffusion counterpart, the electro-diffusion ratio is a thermodynamic quantity. Interestingly, it can then be evaluated in the same relatively simple way as suggested in Ref. [6] for evaluating the baro-diffusion ratio. We outline this calculation in the appendix A.

V. DISCUSSION

Relations (32) and (33) do not provide an explicit result for the electro-diffusion coefficient because of the transport coefficient A_{lh} entering the formula for D. However, as the approach presented does provide an explicit result for k_E, a substantial insight into the role of electro-diffusion can still be gained. To compare baro- and electro-diffusion caused perturbations

of the species concentrations we employ Eqs. (5) and (33) to evaluate the ratio

$$\frac{k_E}{k_p} = \frac{Z_l/m_l - Z_h/m_h}{1/m_l - 1/m_h}, \tag{36}$$

which depends on the properties of the ions only. In the special case of the two isotopes of one element, i.e. $Z_l = Z_h \equiv Z$, Eq. (36) gives $k_E/k_p = Z$. In particular, for the practically important DT mix, the two coefficients turn out to be equal, $k_E/k_p = 1$. For the D^{3}He mix, commonly used to study sub-ignited implosions, $k_E/k_p = -1$. In a plasma shock wave, the electric field is directed towards the unshocked region to prevent electrons' running ahead of ions and maintain quasi-neutrality. It can therefore be observed that in the case of the DT mix baro- and electro-diffusions act together, whereas in the case of the D^{3}He mix the two tend to cancel each other.

Of course, relation between the baro- and electro-diffusion ratios alone is not sufficient for relating the corresponding fluxes. The total ion pressure gradient and the electric field also need to be compared. While carrying out this comparison in a general case is hardly possible, it is reasonable to assume $\nabla \log p_i \sim e\nabla\Phi/T_i$. Moreover, in a shock wave, the electric field is rather governed by the electron pressure gradient. The pressure of electrons is often greater than that of ions and therefore it is likely that the electro-diffusive flux may be noticeably larger than the baro-diffusive flux. This becomes particularly intriguing for the D^{3}He mix, in which electro-diffusion counteracts baro-diffusion. As a result, ^{3}He concentration may be increased over its unperturbed value, contrasting the neutral theory based expectation that it is the lighter fraction whose concentration is enhanced in the shock front [13].

In terms of numerical modeling of the diffusive separation of the fuel ions in ICF capsules, the most direct approach would be to solve the multi-component plasma equations in its individual species form. The electric field is then explicitly evolved. Alternatively, the ion fluid equations can be solved in the center of mass frame, i.e. $\rho, \mathbf{u}, p_i$, with the ion species concentration c followed by Eq. (1) and Eq. (31). With this approach, the electric field can be either independently evolved using Maxwell's equations, or inferred from the equation of motion for the electrons in the quasineutral regime. In this latter case, the electron inertia and electron viscosity are ignored, so

$$e\nabla\Phi = \frac{\nabla p_e}{n_e} - (B_{le} + B_{he})\nabla T_e. \tag{37}$$

The above equation implies that, at a minimum, the fluid equations should evolve the

electron temperature separately from the ions', which fortunately is frequently done in ICF codes.

Finally, we comment on whether or not electro-diffusion, described here by considering the ion species separately, can be attributed to baro-diffusion in the plasma as a whole. As previously mentioned, the plasma mass flux is essentially equal to the ion mass flux, because the electron inertia is negligible. The question to be answered is therefore whether or not the right side of Eq. (31) can be represented in terms of the total plasma pressure gradient, rather than in terms of the partial ion pressure gradient and the electric field. To investigate the issue, we insert Eq. (37) into Eq. (31) to obtain

$$\mathbf{i}_l = -\rho D\Big[\nabla c + \frac{k_p}{(n_l+n_h)T_i}\nabla p_i + \frac{k_E}{n_e T_i}\nabla p_e + k_T^{(i)}\nabla \log T_i + \tilde{k_T}^{(e)}\nabla \log T_e\Big], \tag{38}$$

where

$$\tilde{k_T}^{(e)} = k_T^{(e)} - (B_{le} + B_{he})(T_e/T_i)k_E. \tag{39}$$

Eliminating both the electron and ion partial pressure gradients in Eq. (38) by substituting the total pressure gradient is only possible if

$$\frac{k_E}{k_p} = \frac{n_e}{n_l + n_h} \tag{40}$$

for any values of n_l and n_h.

Combining Eq. (36) and the quasi-neutrality condition $n_e = Z_l n_l + Z_h n_h$, one finds that Eq. (40) may be identically satisfied only for $Z_l = Z_h \equiv Z$. The right side of Eq. (40) is then equal to Z and indeed matches the left side of Eq. (40) according to Eq. (36). Employing $n_e/(n_l + n_h) = k_E/k_p = Z$ in Eq. (38) we find

$$\mathbf{i}_l = -\rho D\Big[\nabla c + \tilde{k_p}\nabla \log p + k_T^{(i)}\nabla \log T_i + \tilde{k_T}^{(e)}\nabla \log T_e\Big], \tag{41}$$

where $p = p_i + p_e$ is the total plasma pressure and

$$\tilde{k_p} = (1 + ZT_e/T_i)k_p. \tag{42}$$

Equation (36) predicts a larger baro-diffusion coefficient, as compared to the case of a neutral binary mix. In the limiting case of $T_e = T_i$

$$\tilde{k_p}(T_e = T_i) = (1 + Z)k_p,$$

giving that the enhancement factor due to electro-diffusion is $(1+Z)$. This factor is familiar from the well-known ambipolar enhancement for ion diffusion with respect to the laboratory frame. However, here the impact is on relative diffusion of two distinct ion species.

In summary, representing the electric field effect on inter-ion-species diffusion as a modification to the conventional baro-diffusion coefficient is only possible when these ion species are in the same charge state ($Z_l = Z_h$). Moreover, even in such a case, the electron and ion temperatures need to be evolved separately for this effect to be properly accounted for.

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Appendix A: Evaluating electro-diffusion ratio in the Zel'dovich-Raizer fashion

In Ref. [5] expression (5) for the baro-diffusion ratio is obtained by utilizing a general formula giving k_p in terms of the specific volume and chemical potential derivatives over concentration. Instead, Zel'dovich and Raizer [6] notice that once k_p is known to be a thermodynamic quantity, the answer found in some special case should also work for all other cases. In particular, evaluating k_p can be simplified by considering a globally equilibrated system. Indeed, the flux, as well as the temperature gradient, is then equal to zero and the baro-diffusion ratio can be obtained by balancing the ∇c and $\nabla \log p$ terms on the right side of Eq. (4). By writing explicit expressions for the densities of the mix components in the uniform gravitational field Eq. (5) can then be recovered. Below, we apply this idea to a plasma with two sorts of ions.

First, we recall Eq. (6) and set $\nabla T_i = 0 = \mathbf{i}_{l,h}$ to obtain

$$\frac{dc}{dx} + k_p \frac{d \log p_i}{dx} + \frac{e k_E}{T_i} \frac{d\Phi}{dx} = 0, \tag{A1}$$

where k_p is readily provided by Eq. (5). Next, we notice that for a plasma equilibrated in the uniform gravitational field, the light and heavy ion density profiles are given by

$$n_{l,h} = n_{l0,h0} \exp\Big(-\frac{m_{l,h} g x}{T_i} - \frac{Z_{l,h} e \Phi}{T_i}\Big), \tag{A2}$$

where $n_{l0,h0}$ are the species number densities at $x = 0$ and $\Phi(x = 0) = 0$. Now, the first two terms on the left side of Eq. (A1) need to be evaluated with the help of Eq. (A2).

To do so, we observe that $c = m_l n_l/(m_l n_l + m_h n_h)$ to write

$$\frac{dc}{dx} = -c^2 \frac{d}{dx}\Big(\frac{1}{c}\Big) = -c^2 \frac{d}{dx}\Big(\frac{m_h n_h}{m_l n_l}\Big). \tag{A3}$$

Next, we insert Eq. (A2) into the right side of Eq. (A3) to find

$$\frac{dc}{dx} = -c(1-c)\Big[-\frac{g}{T_i}(m_h - m_l) + \frac{eE}{T_i}(Z_h - Z_l)\Big]. \tag{A4}$$

Finally, the total ion pressure gradient is calculated along the same lines to obtain

$$\frac{d \log p_i}{dx} = -\frac{g}{T_i} \frac{(m_l n_l + m_h n_h)}{n_l + n_h} + \frac{eE}{T_i} \frac{(Z_l n_l + Z_h n_h)}{n_l + n_h}. \tag{A5}$$

Then, by inserting Eqs. (5), (A4) and (A5) into Eq. (A1) and solving it for k_E, previously obtained result (33) is reproduced.

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[1] If instead the laser driver rather than the target temperature is held fixed, optimal fusion yield can be obtained with different reactant number densities which leads to a higher target temperature upon implosion, espcially in sub-ignition plasmas.

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